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Supplemental Material

Estimating Inorganic Arsenic Exposure from U.S. Rice and Total Water Intakes

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Censoring of the Second Six Year Review Database for Arsenic in Drinking Water

U.S. Environmental Protection Agency's (U.S. EPA) Drinking Water Database. In this study, the Second Six-Year Review database from the Office of Ground Water and Drinking Water (U.S. EPA 2010) was used to estimate the arsenic concentrations available to the U.S. consumer. This database includes 224,035 records from 49,473 drinking water utilities in 45 states collected from 1998−2005. Of the 224,035 records for arsenic, 141,357 records were labeled as no detects. Of the 141,357 no detects, 95,163 records reported a detection limit of ≤ 2 ppb and 134,198 records reported a detection limit ≤ 5 ppb. The authors did not conduct a record-by-record evaluation, but some anomalies associated with concentration units used to report the data seemed apparent. Three of these anomalies are listed below leading to two different censoring criteria.

1. Utilities reported no detects greater than 10 ng/mL. Censoring: If the utilities reported no detect concentrations greater than 10 ng/mL, then concentrations were set equal to 10 ng/mL (affected 740 individual records of 141,357 no detects; range of 10.1−5,000 ng/mL).
2. Utilities reported no detects less than 0.5 ng/mL. Censoring: no detects that were less than 0.5 ng/mL were set equal to 0.5 ng/mL (affected 881 individual records of 141,357 no detects; range of 0.001−0.49 ng/mL).
3. Detects over 100 ng/mL were examined per utility and sample point identification (ID) to determine whether concentration was an outlier. There were 324 records from 98 utilities that were found to be greater than 100 ppb. Forty-seven of the utilities had more than one entry that exceeded 100 ng/mL, and for this reason, concentrations were left as reported. Fifty-one utilities were individually examined and 16 were found to have potential data quality inconsistencies. Censoring: none.

The first criteria attempts to minimize the impact of utilities reporting no detect concentrations above the least sensitive compliance monitoring spectroscopic technique (EPA Method 200.7). EPA Method 200.7 has a drinking water MDL of 8 ng/mL. In some cases, the utility seemed to report the concentration in micrograms per milliliter, while the appropriate units were nanograms per milliliter. In one case, the utility reported 5 µg/mL as a no detect concentration, and because of the first censoring criteria, this no detect was set to 10 ng/mL. The second censoring criteria attempted to minimize the impact of utilities that reported no detects at extremely low concentration. Again, inappropriate units used in reporting the concentrations seemed to be the cause with one utility reporting a detection limit of 1 part per trillion. Finally, elevated drinking water concentrations were evaluated on a case-by-case basis, and the conclusion after this detailed evaluation was to not censor any of the detected concentrations reported in the database.

No detects represent the majority (141,357/224,035) of the records in the censored data set. One-half the reported detection limit was assigned to all no detect records when calculating the best estimate for the arsenic concentration for that particular utility. The population-weighted geometric mean is 1.8 ng/mL with a geometric standard deviation of 2.5. A sensitivity analysis was conducted in which no detects were replaced with ⅛, ¼, ½, ¾, and 1 MDL and the resulting cumulative density functions for ⅛, ½, and 1 MDL are included in Figure 4A (U.S. population) and Supplemental Material, Figure S5 (1 – 2 year old U.S. subpopulation).

Finally, by incorporating this most recent drinking water data set into the exposure model, a more complete representation of the iAs exposure is possible. According to this database, 2,486 utilities, serving 4.6 million people, had an average iAs concentrations above 10 ng/mL with 0.1 million above 40 ng/mL in 2005. A number of these utilities may have installed treatment after 2005, so this database overestimates the arsenic concentration associated with these utilities.

Rice Sampling Protocol

Rice mill sampling and production-weighted composite protocols. Each participating mill was sent a set of color-coded, grain-specific, sampling containers and a set of instructions. The grain-specific samples were collected once a week over an 8-wk period by an employee at each of the 21 mills. The sampler was asked to follow U.S. Department of Agriculture (USDA) protocol for sampling and to sample only rice intended for U.S. markets and to exclude rice intended for use in pet foods or beer production. If the mill did not produce that grain type during a particular sampling week, the sample containers were returned empty. The samples were shipped to an intermediate party to assure individual mills could not be identified and with chain-of-custody documentation to assure the integrity of the samples. These mill- and grain-specific samples were used to make the 40 domestic rice composites.

Table S1. A summary and comparison of databases used in the literature to simulate arsenic exposure in the U.S. population.[a]

	Xue et al. (2010)	Meacher et al. (2002)	Tsuji et al. (2007)	Yost et al. (2004) (1−6 year olds)	Schoof et al. (1999a)
Drinking Water Database	NRDC[b] (1980−1998) 8,970 utilities from 25 states from U.S. EPA AOED	NAOS[c] 500 utilities that serve a population > 1,000	U.S. EPA AOED[d] from 25 states	Not utilized	Not utilized
Drinking Water Ingestion Rate This paper: $\bar{x}$ = 1.54 L/day	No Drinking Water ingestion rate reported	20−64 years of age 1.12 ± 1.63 L/day[e]	Direct: $\bar{x}$ = 635−840 g/day Indirect: $\bar{x}$ = 488−549 g/day[f]	No direct drinking water ingestion rate utilized	No direct drinking water ingestion rate utilized
Drinking Water iAs Exposure This paper: $\bar{x}$ = 4.2 µg iAs/day	U.S. population $\bar{x}$ = 0.025 ± 0.104 µg iAs/kg-day	18−59 age $\bar{x}$ = 2.5 µg iAs/day[g]	U.S. population $\bar{x}$ = 2.5 µg iAs/day (untruncated data set)	0.8 ng iAs/g[h] (Indirect: used in food preparation)	Not utilized
Drinking Water Database Comparison to this paper	The Second Six Year Review is the most current and comprehensive finished drinking water data set from both a population-served and a geographic/utility-coverage perspective. Data for Second Six Year Review were collected from 1998−2005 from 49,473 utilities located in 45 states. By comparison, the NRDC or U.S. EPA AOED data were collected from 1980−1998 from 8,970 utilities located in 25 states. The NAOS collected source water from 500 utilities (pop. served > 1,000) and predicted iAs based on the type of drinking water treatment the utility reported. In addition, new source waters have been identified (approximately 31% of the utilities in the NRDC data set are in the current Second Six Year Review data set), and the iAs levels in old sources may have changed since 1980. Newer analytical procedures are more likely to be included in the Second Six Year Review, thereby improving data quality and reporting limits.			Yost et al. (2004) focused on dietary exposure estimates and used 0.8 ng iAs/g to estimate the indirect water exposure from food preparation.	Schoof et al. (1999a) focused on dietary exposure estimates and did not utilize a drinking water database.
Dietary TAs Database	FDA's Total Dietary Survey 1991−2004 (280 core foods)[i]	Not utilized	Not utilized	Not utilized	Not utilized
Dietary iAs Database	Schoof et al. (1999b) (40 dietary commodities from 2 locations)[j]			Schoof et al. (1999b) (38 dietary commodities from 2 locations)[j]	Schoof et al. (1999b) (40 dietary commodities from 2 locations)[k]
Database: Dietary/Tapwater Ingestion Rate	National Health and Nutrition Examination Survey 2003−2004[l] 16,934 person-days Recipe files from the Food Commodity Intake Database	CSFII 1992−1994[m] 16,821 daily records Recipe files from USDA 1989−1991	CSFII 1994−1996 U.S. population, 1998 Supplemental Children's Survey, recipe files from Exponent's FARE™		CSFII 1989−1992
Estimated Dietary TAs Exposure	U.S. population 0.36 ± 1.28 µg TAs/kg-day	Not utilized	Not utilized	Not utilized	Not utilized
Estimated Dietary iAs Exposure This paper: $\bar{x}$ = 1.4 µg iAs/day	U.S. population $\bar{x}$ = 0.05 ± 0.09 µg iAs/kg-day Rice: $\bar{x}$ = 0.6 µg/day	18−59 age $\bar{x}$ = 3.1 µg iAs/day[g]	U.S. population $\bar{x}$ = 3.6 µg iAs/day Rice: $\bar{x}$ = 1 µg/day[n]	1−6 year olds $\bar{x}$ = 3.2 µg iAs/day Rice: $\bar{x}$ = 0.6 µg/day	18−59 age $\bar{x}$ = 3.2 µg iAs/day
Dietary Speciation Database Comparison to this paper	This paper focuses on rice, a single dietary commodity, because rice is considered one of the major (~20%) dietary exposure sources for iAs. Mantha et al. estimated the distribution of iAs in rice available to the U.S. consumer and used in the SHED's exposure model. They utilized a production-weighted sampling of grain types collected over an 8-week period. The grain types were analyzed by both a quantitative speciation approach, which determines all the iAs in the grain, and by a Gastrointestinal-based extraction, which provides a bioaccessibility estimate for the IAs in each grain type. Other studies to date have adopted a constant iAs/TAs ratio (Schoof et al. 1999b) in rice and other commodities.				

Abbreviations: AOED, Arsenic Occurrence and Exposure Dataset; CSFII, Continuing Survey of Food Intakes by Individuals; FARE, Food and Residue Evaluation; FDA, Food and Drug Administration; iAs, inorganic arsenic; ICP-MS, inductively coupled plasma mass spectrometry; MDL, method detection limit; NAOS, National Arsenic Occurrence Survey; NRDC, National Resource Defense Council; OME, Ontario Ministry of the Environment; TA, total arsenic; USDA, U.S. Department of Agriculture; U.S. EPA, U.S. Environmental Protection Agency; $\bar{x}$, mean.

[a]Yost et al. (1998) also utilize dietary speciation databases to estimate iAs exposures but rely on an even older dietary speciation database collected by the OME, Report No. 87-48-45000-057: Organic vs. Inorganic Arsenic in Selected Food Samples Toronto, Ontario, Canada, Ontario Ministry of the Environment, Hazardous Contaminants Coordination Branch, 1987. [b]The NRDC data set was derived from the U.S. EPA AOED (U.S. EPA 815-R-00-023); it contains arsenic concentrations from 8,970 utilities from 25 states and is derived from a voluntary data call covering 1980 to 1998 to support the arsenic rule. NRDC replaced no detects with ½ MDL and has censored the AOED data set as described on its website (http://www.nrdc.org/water/drinking/arsenic/appa.asp [accessed April 29, 2014]). Xue et al. (2010) population weight the NRDC data set and calculate a geometric mean and standard deviation of 1.03 ± 4.06 ng iAs/g after replacing all the no detects with ½ MDL. [c]The arsenic concentrations for 500 surface and ground water sources used for drinking water supplies were determined by ICP-MS (~0.1 ng iAs/g detection limit), and then the arsenic concentration was predicted by applying the most likely treatment removal strategy for that system. Neighboring states' arsenic estimates were used to infer arsenic concentration for states when data were unavailable. [d]U.S. EPA AOED (U.S. EPA 815-R-00-023) was collected from 1980−1998 from 25 states. [e]Geometric mean and standard deviation of 1.122 L/day ± 1.63 from tap water (consumed directly as a beverage or used to prepare food) for 20−64-year-old individuals. This data was collected from 11,731 observations and corresponds to an arithmetic mean of 1.265 L/day. These values were initially estimated by Ershow and Cantor (1989) and summarized in the *Exposure Factors Handbook,* September 2011. [f]These ranges were calculated using four U.S. regions with the southwest reporting the highest consumption of tap water. The data can be converted to mL/day by using the density of water equals 1 g/mL. [g]This mean is calculated from the paper by equally weighting the independent exposure estimates for males and females aged 18−59 years. [h]The arsenic in tap water was collected from two locations and used to estimate the arsenic concentration (0.8 ng iAs/g) in the water used in food preparation. [i]If a commodity's concentration is always reported as <MDL within the database, then that commodity's As concentration is set to zero; but, if any commodity sample in the database is >MDL, then all no detects for that commodity are assumed to be equal to ½ MDL. [j]The Schoof Market Basket Survey collected four samples each of 40 commodities from two Texas communities in 1997. Inorganic arsenic was undetectable in approximately one-half of the commodity samples. In some cases, it was necessary to infer the iAs concentration for some commodities. For example, the average iAs in all fruits was used to estimate the iAs in pineapple, mango, and raspberries. Extrapolation process referenced to 40 CFR 180.41. The arsenic concentration in tap water (0.8 ng iAs/g) used for food preparation was estimated from samples collected from two locations. [k]The Schoof Market Basket Survey collected four samples each of 40 commodities from two Texas communities in 1997. Inorganic arsenic was undetectable in approximately one-half of the commodity samples. In some cases, it was necessary to infer the iAs concentration for some commodities. For example, the average iAs in all fruits were used to estimate the iAs in pineapple, mango, and raspberries. Extrapolation process referenced to 40 CFR 180.41. [l]U.S. EPA Food Commodity Intake Database contains recipes identifying the raw agricultural commodities typically comprising foods identified as consumed in National Health and Nutrition Examination Survey (n = 16,934 person-days, dietary ingestions instantly recorded). Total water consumption equals direct plus indirect using a single source of water. [m]CSFII 1992−1994, 16,821 dietary records for 3 consecutive days for 18−59-year-old individuals living in the United States. [n]This exposure is estimated based on inferring a concentration from a graph.

Table S2. Total and speciated arsenic for U.S. consumption weighted composites of imported rice using a mass balance approach after a DNAS.[a,b]

Imported Rice		Total Digest[e]		As Extraction Totals[f]		Average Extraction Efficiency[g] (% Rec)	As Speciation in DNAS[h]							Average Chrom Recovery[i] (% Rec)
							iAs		DMA		MMA		Sum of Speciation ($As_{\Sigma Chrom}$) (ng/g)	
Rice Type[c]	RWF[d]	Conc (As_{TD}) (n=3) (ng/g ± 2σ)	LFM (As_{TD}) (% Rec)	Conc (As_{TE}) (ng/g ± 2σ)	LFM (As_{TE}) (% Rec)		Conc (ng/g ± 2σ)	LFM_{iAs} (n=1) (% Rec)	Conc (ng/g ± 2σ)	LFM_{DMA} (n=1) (% Rec)	Conc (ng/g ± 2σ)	LFM_{MMA} (n=1) (% Rec)		
TLG-OH-01		149.9 ± 16.2	98	154.1 ± 6.4	99.2	102.8	98.0 ± 1.7	100.2	52.5 ± 1.7	100.9	2.4 ± 0.6	102.6	152.9	99.2
IB-OH-01		80.2 ± 18.4	99	90.2 ± 4.5	95.9	112.5	71.3 ± 4.0	104.1	22.0 ± 0.5	112.0	6.3 ± 1.7	109.1	99.6	110.5
TLG-NY-02		190.3 ± 24.8	99	167.0 ± 1.6	106.9	87.8	101.5 ± 0.8	107.4	71.2 ± 1.9	123.5	7.6 ± 1.3	111.1	180.3	108.0
IB-NY-02		59.7 ± 2.7	100	59.2 ± 3.0	100.3	99.2	43.3 ± 0.9	111.4	19.1 ± 0.6	97.8	ND	112.7	62.4	105.4
IB-NJ-03		56.6 ± 11.4	105	51.1 ± 2.7	97.9	90.4	39.9 ± 0.7	110.6	13.8 ± 1.0	100.4	ND	109.2	53.7	105.1
TLG-NJ-03		151.3 ± 3.8	86	152.9 ± 12.0	100.1	101.1	90.8 ± 8.6	105.7	60.4 ± 8.2	99.8	3.3 ± 0.6	107.5	154.6	101.1
TLG-FL-04		107.0 ± 1.6	96	97.3 ± 4.4	100.5	91.0	74.1 ± 1.5	106.7	27.7 ± 3.6	106.7	ND	120.3	101.9	104.7
IB-FL-04		54.4 ± 3.6	97	50.5 ± 4.7	99.0	92.8	30.2 ± 2.8	103.2	21.1 ± 0.1	104.5	ND	109.3	51.3	97.4
IB-AZ-05		109.8 ± 8.8	100	109.9 ± 3.3	93.5	100.1	83.3 ± 3.0	103.0	24.6 ± 2.0	97.3	ND	115.7	108.0	98.2
TLG-AZ-05		173.9 ± 1.8	103	158.5 ± 24.5	101.2	91.2	98.2 ± 7.4	115.8	52.0 ± 9.1	112.5	3.3 ± 1.0	112.9	153.4	97.0
TLG-NC-06		183.2 ± 27.6	107	162.7 ± 8.0	104.8	88.8	115.7 ± 7.8	110.8	52.4 ± 5.2	106.9	2.8 ± 0.3	104.3	170.9	105.1
IB-NC-06		86.1 ± 7.1	98	77.3 ± 1.5	102.1	89.8	62.3 ± 4.2	111.1	18.7 ± 1.6	106.2	ND	113.6	81.0	104.8
TLG-WA-07		144.0 ± 8.0	98	143.1 ± 4.1	100.8	99.3	93.9 ± 2.2	107.2	49.7 ± 1.6	103.6	3.0 ± 0.6	121.7	146.6	102.5
IB-WA-07		94.0 ± 6.32	101	94.1 ± 2.1	104.3	100.1	66.6 ± 0.1[j]	108.4	26.1 ± 1.1[j]	108.3	2.6 ± 1.3[j]	121.7	95.2	100.6
Across Matrix Avg[k] ± 2σ		**117.2 ± 95.8**	**99.1 ± 9.7**	**112.0 ± 87.1**	**100.4 ± 7.0**	**96.2 ± 14.1**	**76.4 ± 51.3**	**107.5 ± 8.3**	**36.5 ± 37.7**	**105.7 ± 14.0**	**3.9 ± 3.9**	**112.3 ± 12.0**	**115.1 ± 88.5**	**102.8 ± 8.2**
SRM[l] 1568a ($\bar{x}$ ± 2σ)		**307 ± 17.4**		**294.7 ± 19.2**		**101.6 ± 6.6**	**102.1 ± 4.0**		**172.7 ± 10.2**		**13.4 ± 6.1**		**288.2 ± 16.1**	**97.8 ± 3.3**

Abbreviations: As_{TD}, the total digested arsenic; As_{TE}, total extracted arsenic; $As_{\Sigma Chrom}$, the sum of the individual arsenic species in a rice composite; DMA, dimethylarsinic acid; DNAS, dilute nitric acid extraction; iAs, inorganic arsenic; IB, Indian basmati rice; LFB, laboratory-fortified blank; LFM, laboratory-fortified matrix; MMA, monomethylarsonic acid; ND, no detect; RWF, relative weighting factor; SRM, standard reference material; TLG, Thailand long grain rice.

[a]All analyses are reported based on oven-dried sample weights. All concentrations (ng/g) are reported as averages ($\bar{x}$ ± 2σ, *n* = 3 replicates from cooked rice sample) except where noted. [b]Two types of total arsenic LFBs were analyzed. The first was a LFB As_{TD} and the second was a LFB As_{TE}. The LFB As_{TD} was 100 ± 8% and the LFB As_{TE} was 100 ± 2%. The samples were fortified prior to digestion with 4.8 to 8 ppb As for the As_{TD} LFB and LFM. Two types of species-specific arsenic LFBs were analyzed. The first was an LFB for the iAs (LFB_{iAs}) and the second was an LFB for DMA (LFB_{DMA}). The LFB_{iAs} was 101 ± 4% and the LFB_{DMA} was 101 ± 4%. The LFB extracts were fortified with 6 ppb iAs, 4 ppb DMA, and 2 ppb MMA. [c]Samples were categorized according to U.S. Rice Federation Production classifications. The rice grain type is followed by OH, NY, NJ, FL, AZ, NC, WA, representing the state in which the samples were collected. [d]The production data were not available for imports, and for this reason, the U.S. Rice Federation 2008 Domestic Usage Report (USA Rice Federation 2008) was used to estimate the total amount of imported rice, while the website at the U.S. Department of Agriculture, Foreign Agricultural Service, Global Agricultural Trade System Online (USDA 2015b) was used to further delineate this percentage to the country of origin. [e]As_{TD} is a total arsenic determination after a hot mineral acid dissolution. LFM As_{TD} is an LFM on a sample that will undergo a hot mineral acid dissolution. [f]As_{TE} is a flow injection total arsenic determination in the extraction fluid. As_{TE} is determined using a single-point method of standard addition. LFM As_{TE} is a LFM on the total arsenic in the extraction fluid. [g]Extraction efficiency equals the As_{TE} in a rice composite, divided by the total arsenic after a hot mineral acid dissolution (As_{TD}) times 100. [h]The LFM extract was fortified with 6 ppb iAs, 4 ppb DMA, and 2 ppb MMA. Inorganic arsenic (iAs = As^{III} + As^{V}). LFM_{iAs} is a species-specific LFM for iAs in the extraction fluid. LFM_{DMA} is a species-specific LFM for DMA in the extraction fluid. LFM_{MMA} is a species-specific LFM for MMA in the extraction fluid. [i]Average chromatographic recovery equals $As_{\Sigma Chrom}$ divided by As_{TE} for that rice composite times 100. [j]These concentrations and percentages were determined using *n* = 2. [k]Across-matrix average and sigma are calculated using the mean for each rice composite and do not include data from the SRM. The across-matrix average and sigma are arithmetic not production weighted. [l]The mass balance terms have been calculated for the SRM by pooling the data from individual batches (*n* = 14).

Table S3. Total and speciated arsenic for U.S. consumption weighted composites of domestic rice using a mass balance approach after a DNAS.[a,b]

Domestic Rice		Total Digest[e]		As Extraction Totals[f]		Average Extraction Efficiency[g] (% Rec)	As Speciation in DNAS[h]							Average Chrom Recovery[i] (% Rec)
							iAs		DMA		MMA		Sum of Speciation ($As_{\Sigma Chrom}$) (ng/g)	
Rice Type[c]	RWF[d]	Conc (As_{TD}) (n=3) (ng/g ± 2σ)	LFM (As_{TD}) (% Rec)	Conc (As_{TE}) (ng/g ± 2σ)	LFM (As_{TE}) (% Rec)		Conc (ng/g ± 2σ)	LFM_{iAs} (n=1) (% Rec)	Conc (ng/g ± 2σ)	LFM_{DMA} (n=1) (% Rec)	Conc (ng/g ± 2σ)	LFM_{MMA} (n=1) (% Rec)		
Ins	0.8	87.9 ± 8.0	93	94.5 ± 7.7	74.1	107.5	43.2 ± 1.4	83.9	50.3 ± 2.9	69.4	2.3 ± 0.7	108.1	95.8	101.4
Ins	0.8	122.8 ± 5.6	111	137.3 ± 9.6	106.1	111.8	55.0 ± 1.2	106.3	82.9 ± 3.6	99.0	3.3 ± 0.1	102.8	141.3	102.9
Ins	0.8	109.2 ± 18.8	98	113.5 ± 6.7	90.2	103.9	50.8 ± 0.8	108.4	65.2 ± 1.6	104.2	2.9 ± 0.4	114.6	118.9	104.8
Ins	0.8	110.7 ± 10.8	103	111.2 ± 1.8	99.2	100.4	55.0 ± 0.9	110.3	56.8 ± 2.3	101.2	2.1 ± 0.8	123.0	113.9	102.5
Par	4.4	242.0 ± 16.0	111	221.3 ± 19.7	90.9	91.4	134 ± 5.4	113.3	91.4 ± 4.4	105.3	4.2 ± 0.4	122.2	229.5	103.8
Par	4.4	236.4 ± 38.8	109	225.5 ± 11.6	101.3	95.4	121.8 ± 2.8	97.3	93.2 ± 0.8	101.7	4.1 ± 0.6	108.2	219.1	97.2
Par	3.4	243.1 ± 15.0	108	218.7 ± 10.3	117.0	90.0	131.4 ± 2.5	108.9	88.3 ± 7.4	109.2	3.2 ± 0.5	105.2	222.8	101.9
Par	4.6	227.5 ± 45.0	105	217.2 ± 8.0	107.1	95.5	125.4 ± 3.9	105.1	89.5 ± 0.7	105.1	4.3 ± 0.1	101.2	219.2	100.9
WSG	0.1	101.7 ± 6.0	99	105.5 ± 1.9	97.2	103.8	87.8 ± 3.0	101.9	25.1 ± 1.4	101.1	ND	109.3	112.9	107.0
WSG	0.3	142.0 ± 6.4	100	135 ± 4.7	99.9	95.1	105.2 ± 6.3	120.0	36.9 ± 2.8	108.0	ND	113.9	142.1	105.7
WSG	0.0	157.9 ± 12.0	100	154.3 ± 3.5	99.3	97.7	75.6 ± 3.2	101.4	83 ± 0.8	107.5	2.2 ± 1.0	109.9	160.7	104.2
WSG	0.0	132.9 ± 4.8	98	126.9 ± 2.0	100.3	95.5	98.3 ± 1.2	105.7	35.9 ± 0.8	104.6	ND	103.9	134.2	105.7
Brn	0.2	190.2 ± 17.2	110	174.2 ± 4.7	110.7	91.6	143.5 ± 5.3	107.1	40.5 ± 2.5	115.8	ND	106.4	184.0	105.6
Brn	0.5	222.5 ± 22.0	95	186.7 ± 6.0	96.4	83.9	157.6 ± 2.6	105.9	37.8 ± 2.3	101.3	2.0 ± 0.9	103.6	197.4	105.7
Brn	0.2	118.3 ± 8.0	88	125.6 ± 3.9	76.3	106.1	92.1 ± 1.4	72.9	36.9 ± 3.1	87.8	ND	104.8	129.0	102.8
Brn	0.3	211.3 ± 11.8	88	183.3 ± 3.2	104.1	86.7	147.5 ± 1.4	111.0	45.6 ± 2.5	111.4	2.1 ± 0.2	104.7	195.1	106.5
Brn	0.7	262.6 ± 22.8	96	263.7 ± 10.6	97.3	100.4	154.9 ± 4.7	106.6	105.9 ± 1.7	106.0	4.5 ± 1.0	106.5	265.2	100.6
Brn	1.0	227.4 ± 11.6	100	217.3 ± 5.4	104.6	95.6	119.6 ± 6.6	107.1	96 ± 1.4	105.1	4.1 ± 0.4	106.7	219.7	101.1
Brn	0.6	260.4 ± 21.0	103	270.8 ± 14.7	105.6	104.0	151.9 ± 4.3	112.8	121.6 ± 0.7	112.2	5.6 ± 0.6	106.5	279.2	103.1
WMG	1.0	99.1 ± 4.4	108	105.2 ± 4.1	89.6	106.1	71.8 ± 0.7	105.5	38 ± 1.6	105.4	ND	111.5	109.8	104.5
WMG	3.5	118.8 ± 2.6	103	115.2 ± 3.4	97.5	96.9	77 ± 2.6	106.9	43.7 ± 1.7	106.0	ND	107.6	120.7	104.8
WMG	0.7	181.4 ± 3.2	101	178.1 ± 7.5	102.4	98.2	97 ± 2.2	108.6	85.4 ± 1.0	105.0	ND	115.5	182.4	102.4
WMG	3.7	166.4 ± 17.2	95	168.1 ± 5.9	104.4	101.0	86.7 ± 1.0	106.8	83.7 ± 3.5	106.2	2.0 ± 0.1	107.5	172.4	102.6
WMG	0.7	176.1 ± 16.0	103	145.8 ± 3.4	104.8	82.8	75.8 ± 2.7	104.9	75.9 ± 3.1	108.8	5.2 ± 0.7	97.2	156.9	107.6
WMG	6.4	73.8 ± 10.0	99	70.9 ± 6.4	100.5	96.0	63.5 ± 3.7	102.7	13.5 ± 5.6	111.9	4.8 ± 0.9	104.9	81.8	115.4
WMG	6.4	48.0 ± 2.8	96	51.3 ± 1.0	98.4	106.8	43.7 ± 1.1	105.3	10.3 ± 0.5	104.5	ND	107.7	54.0	105.3
WMG	1.2	195.9 ± 16.4	99	192.1 ± 11.3	97.3	98.0	100.2 ± 1.2	109.2	96.2 ± 0.8	104.6	2.6 ± 0.2	105.9	199.0	103.7
WLG	3.2	183.1 ± 3.4	93	175.4 ± 8.0	90.7	95.8	101.2 ± 1.3	97.6	80.6 ± 1.1	94.5	2.6 ± 0.2	96.7	184.4	105.1
WLG	3.2	210.9 ± 32.4	113	193.5 ± 2.5	96.9	91.7	103.4 ± 1.8	113.4	99.3 ± 1.5	109.0	2.8 ± 0.1	97.4	205.5	106.2
WLG	3.2	178.8 ± 14.2	109	175.6 ± 6.9	99.4	98.2	92.4 ± 1.1	107.1	81.7 ± 1.1	103.9	2.5 ± 0.4	109.5	176.6	100.6
WLG	3.2	181.9 ± 26.4	115	178.9 ± 4.9	106.4	98.4	90.4 ± 1.0	109.9	92.1 ± 2.5	106.7	2.5 ± 0.5	110.7	184.9	103.4
WLG	0.9	228.7 ± 10.4	96	232.8 ± 10.7	103.7	101.8	132.7 ± 2.1	108.1	113.6 ± 2.6	109.4	3.1 ± 1.4	109.9	249.4	107.1
WLG	8.0	186.2 ± 11.0	100	196.7 ± 5.0	103.3	105.7	98.4 ± 0.5	109.0	108.9 ± 1.7	100.2	2.6 ± 0.3	109.4	209.9	106.7
WLG	4.9	185.9 ± 10.2	97	195.2 ± 7.7	107.0	105.0	91 ± 2.4	104.3	103.8 ± 3.8	96.9	2.9 ± 0.2	112.3	197.7	101.3
WLG	5.7	166.7 ± 2.6	93	175.3 ± 2.8	105.3	105.2	82.8 ± 3.0	106.7	92.1 ± 1.5	102.1	2.9 ± 1.3	110.6	177.7	101.4
WLG	2.0	202.4 ± 25.4	88	206.8 ± 13.2	92.7	102.2	78.4 ± 5.6	88.4	114.4 ± 1.0	103.5	3.7 ± 0.0	106.0	196.4	95.1
WLG	5.2	151.6 ± 16.0	101	148.1 ± 4.7	97.8	97.7	75.9 ± 1.9	92.1	67.6 ± 1.9	120.7	3.1 ± 0.3	114.1	146.5	99.0
WLG	7.1	151.1 ± 10.6	100	165.5 ± 26.3	97.0	109.5	82.3 ± 1.8[j]	102.9	82.9 ± 1.9[j]	105.5	3.8 ± 1.0[j]	122.7	169.0	102.5
WLG	4.9	276.6 ± 22.2	82	298.8 ± 8.9	92.9	108.0	129.7 ± 3.2	112.9	171.5 ± 4.1	111.4	6.4 ± 0.7	124.3	307.7	103.0
WLG	1.0	284.7 ± 20.6	99	256.4 ± 6.4	99.2	90.1	115.5 ± 4.1	103.2	153.7 ± 2.1	96.4	6.0 ± 0.7	109.5	275.2	107.4
Across Matrix Avg[k] ± 2σ		**176.4 ± 116.5**	**100.1 ± 14.7**	**172.7 ± 110.4**	**99.1 ± 16.0**	**98.8 ± 13.8**	**98.5 ± 62.4**	**104.8 ± 16.8**	**77.3 ± 70.7**	**104.2 ± 16.2**	**3.4 ± 2.4**	**108.8 ± 13.0**	**178.5 ± 112.2**	**103.7 ± 6.7**
SRM[l] 1568a ($\bar{X}$ ± 2σ)		**299.1 ± 31.2**		**285.0 ± 13.0**		**98.3 ± 4.5**	**99.7 ± 4.2**		**168.7 ± 11.5**		**11.9 ± 3.3**		**280.3 ± 14.5**	**98.4 ± 5.3**

Abbreviations: As_{TD}, the total digested arsenic; As_{TE}, total extracted arsenic; $As_{\Sigma Chrom}$, the sum of the individual arsenic species in a rice composite; Brn, brown rice; DMA, dimethylarsinic acid; DNAS, dilute nitric acid extraction; iAs, inorganic arsenic; Ins, instant rice; LFB, laboratory-fortified blank; LFM, laboratory-fortified matrix; MMA, monomethylarsonic acid; ND, no detect; Par, parboiled rice; RWF, relative weighting factor; SRM, standard reference material; WLG, white long grain rice; WMG, white medium grain rice; WSG, white short grain rice.

[a]All analyses are reported based on oven-dried sample weights. All concentrations (ng/g) are reported as averages ($\bar{x} \pm 2\sigma$, $n = 3$ replicates from a composited cooked rice sample) except where noted. [b]Two types of total arsenic LFBs were analyzed. The first was a LFB As_{TD} and the second was a LFB As_{TE}. The LFB As_{TD} was 96.4 ± 14% and the LFB As_{TE} was 100 ± 4%. The samples were fortified prior to digestion with 4.8 to 8 ppb As for the As_{TD} LFB and LFM. Two types of species-specific arsenic LFBs were analyzed. The first was a LFB for the iAs (LFB_{iAs}) and the second was a LFB for DMA (LFB_{DMA}). The LFB_{iAs} was 100 ± 5% and the LFB_{DMA} was 100 ± 7%. The LFB extracts were fortified with 6 ppb iAs, 4 ppb DMA, and 2 ppb MMA. [c]Samples were categorized according to U.S. Rice Federation Production classifications. [d]RWF is calculated based on mill- and grain-specific production data relative to all the domestic rice intended for direct human consumption [(mill- and grain-specific production/all production intended for human consumption) × 100]. [e]As_{TD} is a total arsenic determination after a hot mineral acid dissolution. LFM As_{TD} is a LFM on a sample that will undergo a hot mineral acid dissolution. [f]As_{TE} is a flow injection total arsenic determination in the extraction fluid. As_{TE} is determined using a single point method of standard addition. LFM As_{TE} is a LFM on the total arsenic in the extraction fluid. [g]Extraction efficiency equals the As_{TE} in a rice composite, divided by the total arsenic after a hot mineral acid dissolution (As_{TD}) times 100. [h]The LFM extract was fortified with 6 ppb iAs, 4 ppb DMA, and 2 ppb MMA. Inorganic arsenic (iAs = $As^{III} + As^{V}$). LFM_{iAs} is a species-specific LFM for iAs in the extraction fluid. LFM_{DMA} is a species-specific LFM for DMA in the extraction fluid. LFM_{MMA} is a species-specific LFM for MMA in the extraction fluid. [i]Average chromatographic recovery equals $As_{\sum Chrom}$ divided by As_{TE} for that rice composite times 100. [j]These concentrations and percentages were determined using $n = 2$. [k]Across-matrix average and sigma are calculated using the mean for each rice composite and do not include data from the SRM. The across-matrix average and sigma are arithmetic not production weighted. [l]The mass balance terms have been calculated for the SRM by pooling the data from individual batches ($n = 40$).

Table S4. Total and speciated arsenic for U.S. consumption weighted composites of imported rice using a mass balance approach after an *in vitro* gastrointestinal extraction assay.[a,b]

Imported Rice		Total Digest[e]		As Extraction Totals[f]		Average Extraction Efficiency[g] (% Rec)	As Speciation in GI Extract[h]						Average Chrom Recovery[i] (% Rec)
							iAs		DMA		DMMTA	Sum of Speciation ($As_{\sum Chrom}$) (ng/g)	
Rice Type[c]	RWF[d]	Conc (As_{TD}) (n=3) (ng/g ± 2σ)	LFM (As_{TD}) (% Rec)	Conc (As_{TE}) (ng/g ± 2σ)	LFM (As_{TE}) (% Rec)		Conc (ng/g ± 2σ)	LFM_{iAs} (n=1) (% Rec)	Conc (ng/g ± 2σ)	LFM_{DMA} (n=1) (% Rec)	Conc (ng/g ± 2σ)		
TLG-OH-01		149.9 ± 16.2	98	97.4 ± 3.3	79	65	65.8 ± 6.0	83	27.4 ± 2.6	101	ND	93.2	98
IB-OH-01		80.2 ± 18.4	99	44.2 ± 2.6	89	55	31.4 ± 8.7	88	10.4 ± 5.6	99	ND	41.8	115
TLG-NY-02		190.3 ± 24.8	99	114.5 ± 6.3	88	60	37.8 ± 20.3	95	41.3 ± 12.6	99	ND	79.1	90
IB-NY-02		59.7 ± 2.7	100	41.9 ± 1.2	87	70	26.7 ± 1.9	98	13.7 ± 1.4	103	ND	40.4	103
IB-NJ-03		56.6 ± 11.4	105	29.9 ± 5.9	85	53	24.6 ± 0.8	87	11.8 ± 3.7	90	ND	36.4	103
TLG-NJ-03		151.3 ± 3.8	86	109.4 ± 7.1	90	73	62.2 ± 9.1	96	40.4 ± 4.5	90	ND	102.6	98
TLG-FL-04		107.0 ± 1.6	96	71.5 ± 10.7	94	67	53.8 ± 2.8	95	21.9 ± 1.8	100	ND	75.7	110
IB-FL-04		54.4 ± 3.6	97	42.9 ± 21.1	91	79	25.4 ± 14.7	90	13.1 ± 4.4	91	ND	38.5	116
IB-AZ-05		109.8 ± 8.8	100	72.4 ± 12.1	79	66	57.6 ± 1.8	94	13.1 ± 0.8	102	ND	70.7	101
TLG-AZ-05		173.9 ± 1.8	103	93.4 ± 20.2	90	54	65.9 ± 23.7	97	27.8 ± 1.1	102	ND	93.7	100
TLG-NC-06		183.2 ± 27.6	107	113.1 ± 6.3	103	62	85.7 ± 7.5	100	30.4 ± 4.7	93	ND	116.1	100
IB-NC-06		86.1 ± 7.1	98	61.8 ± 8.4	88	72	46.1 ± 6.1	92	8.6 ± 0.8	98	ND	95.4	96
TLG-WA-07		144.0 ± 8.0	98	101.1 ± 12.3	90	70	67.7 ± 24.5	86	27.3 ± 7.0	82	ND	95	100
IB-WA-07		94.0 ± 6.32	101	72.7 ± 7.5	93	77	47.6 ± 2.7	84	18.1 ± 0.3	95	ND	65.7	99
Across Matrix Avg[j] ± 2σ		**117.2 ± 95.8**	**99 ± 10**	**76.2 ± 58.2**	**89 ± 12**	**66 ± 17**	**49.9 ± 37.7**	**92 ± 11**	**21.8 ± 21.7**	**96 ± 12**		**71.7 ± 52.7**	**102 ± 14**
SRM[k] 1568a ($\bar{x}$ ± 2σ)		**307 ± 17.4**		**250.3± 40.1**		**81 ± 15**	**72.5 ± 16.1**		**160.0 ± 22.5**			**264.0 ± 42.0**	**106 ± 20**

Abbreviations: As_{TD}, the total digested arsenic; As_{TE}, total extracted arsenic; $As_{\sum Chrom}$, the sum of the individual arsenic species in a rice composite; DMA, dimethylarsinic acid; DMMTA, dimethylmonothioarsinic acid; iAs, inorganic aresenic; IB, Indian basmati rice; LFB, laboratory-fortified blank; LFM, laboratory-fortified matrix; MMA, monomethylarsonic acid; ND, no detect; RWF, relative weighting factor; SRM, standard reference material; TLG, Thailand long grain rice.

[a]All analyses are reported based on oven-dried sample weights. All concentrations (ng/g) are reported as averages ($\bar{x}$ ± 2σ, *n* = 3 replicates from cooked rice sample) except where noted. [b]Two types of total arsenic LFBs were analyzed. The first was a LFB As_{TD} and the second was a LFB As_{TE}. The LFB As_{TD} was 100 ± 8% and the LFB As_{TE} was 100 ± 8%. The samples were fortified prior to digestion with 4.8 to 8 ppb As for the As_{TD} LFB and LFM. Two types of species-specific arsenic LFBs were analyzed. The first was a LFB for the iAs (LFB_{iAs}) and the second was a LFB for DMA (LFB_{DMA}). The LFB_{iAs} was 101 ± 13% and the LFB_{DMA} was 106 ± 14%. The LFB extracts were fortified with 4 ppb iAs, 3 ppb DMA, and 1.5 ppb MMA. [c]Samples were categorized according to U.S. Rice Federation Production classifications. The rice grain type is followed by OH, NY, NJ, FL, AZ, NC, WA, representing the state in which the samples were collected. [d]The production data were not available for imports and for this reason the U.S. Rice Federation 2008 Domestic Usage Report (USA Rice Federation 2008) was used to estimate the total amount of imported rice, while the website at the U.S. Department of Agriculture, Foreign Agricultural Service, Global Agricultural Trade System Online (USDA 2015b) was used to further delineate this percentage to the country of origin. [e]As_{TD} is a total arsenic determination after a hot mineral acid dissolution. LFM As_{TD} is a LFM on a sample that will undergo a hot mineral acid dissolution. [f]As_{TE} is a flow injection total arsenic determination in the extraction fluid. As_{TE} is determined using a single-point method of standard addition. LFM As_{TE} is a LFM on the total arsenic in the extraction fluid. [g]Extraction efficiency equals the As_{TE} in a rice composite, divided by the total arsenic after a hot mineral acid dissolution (As_{TD}) times 100. [h]The LFM extract was fortified with 4 ppb iAs, 3 ppb DMA, and 1.5 ppb MMA. Inorganic arsenic (iAs = As^{III} + As^{V}). LFM_{iAs} is a species-specific LFM for iAs in the extraction fluid. LFM_{DMA} is a species-specific LFM for DMA in the extraction fluid. LFM_{MMA} is a species-specific LFM for MMA in the extraction fluid. [i]Average chromatographic recovery equals the $As_{\sum Chrom}$ divided by As_{TE} for that rice composite times 100. [j]Across-matrix average and sigma are calculated using the mean for each rice composite and do not include data from the SRM. The across-matrix average and sigma are arithmetic not production weighted. [k]The mass balance terms have been calculated for the SRM by pooling the data from individual batches (*n* = 14).

Table S5. Total and speciated arsenic for U.S. consumption-weighted composites of domestic rice using a mass balance approach after an *in vitro* gastrointestinal extraction assay.[a,b]

Domestic Rice		Total Digest[e]		As Extraction Totals[f]		Average Extraction Efficiency[g] (% Rec)	As Speciation in GI Extract[h]						Average Chrom Recovery[i] (% Rec)
							iAs		DMA		DMMTA	Sum of Speciation	
Rice Type[c]	RWF[d]	Conc (As_{TD}) (n=3) (ng/g $\pm 2\sigma$)	LFM (As_{TD}) (% Rec)	Conc (As_{TE}) (ng/g $\pm 2\sigma$)	LFM (As_{TE}) (% Rec)		Conc (ng/g $\pm 2\sigma$)	LFM_{iAs} (n=1) (% Rec)	Conc (ng/g $\pm 2\sigma$)	LFM_{DMA} (n=1) (% Rec)	Conc (ng/g $\pm 2\sigma$)	($As_{\Sigma Chrom}$) (ng/g)	
Ins	0.793	87.9 ± 8.0	93	79.0 ± 7.4	90	90	32.3 ± 2.9	89	30.7 ± 2.3	89	5.1 ± 0.5	68.1	86
Ins	0.793	122.8 ± 5.6	111	101.1 ± 11.1	84	82	34.8 ± 0.5	87	43.3 ± 2.7	92	9.1 ± 0.6	87.2	87
Ins	0.793	109.2 ± 18.8	98	89.6 ± 14.8	91	82	27.8 ± 1.2	85	36.0 ± 7.3	84	8.4 ± 1.1	72.2	78
Ins	0.793	110.7 ± 10.8	103	88.3 ± 7.9	91	80	35.8 ± 1.1	90	27.3 ± 0.9	86	9.4 ± 0.6	72.5	80
Par	4.447	242.0 ± 16.0	111	182.2 ± 15.9	90	75	91.6 ± 3.5	85	52.2 ± 1.3	84	9.3 ± 0.3	153.1	83
Par	4.363	236.4 ± 38.8	109	169 ± 16.4	105	71	76.1 ± 7.5	93	43.1 ± 8.0	98	18.6 ± 5.6	137.8	80
Par	3.356	243.1 ± 15.0	108	188.3 ± 12.4	89	77	105.4 ± 3.9	95	39.0 ± 4.1	90	14.3 ± 1.8	158.7	86
Par	4.629	227.5 ± 45.0	105	184.8 ± 12.2	91	81	105.3 ± 18.9	87	45.6 ± 4.7	89	9.9 ± 2.3	160.8	88
WSG	0.116	101.7 ± 6.0	99	81.4 ± 8.4	82	80	66.9 ± 4.6	90	18.0 ± 3.1	88	ND	84.9	105
WSG	0.256	142.0 ± 6.4	100	115.3 ± 30.7	86	81	84.3 ± 2.0	90	20.4 ± 1.4	80	ND	104.7	90
WSG	0.012	157.9 ± 12.0	100	104.4 ± 10.7	92	66	50.3 ± 3.1	96	45.2 ± 3.4	92	ND	95.5	91
WSG	0.032	132.9 ± 4.8	98	103.8 ± 4.1	90	78	76.3 ± 4.9	84	19.1 ± 4.9	77	ND	95.4	90
Brn	0.193	190.2 ± 17.2	110	127.9 ± 5.1	95	67	104.6 ± 3.4	97	11.1 ± 0.7	87	ND	115.7	90
Brn	0.539	222.5 ± 22.0	95	145.8 ± 14.8	84	66	127.1 ± 2.8	86	13.6 ± 3.6	83	ND	140.7	92
Brn	0.209	118.3 ± 8.0	88	88.8 ± 6.1	88	75	69.2 ± 1.7	94	12.8 ± 0.5	84	ND	82	93
Brn	0.28	211.3 ± 11.8	88	139.2 ± 10.9	88	66	124.5 ± 6.2	93	13.3 ± 0.2	82	ND	137.8	94
Brn	0.741	262.6 ± 22.8	96	168.4 ± 22.7	93	64	113.0 ± 9.7	90	36.0 ± 2.6	101	ND	149	89
Brn	0.954	227.4 ± 11.6	100	143.3 ± 16.5	96	63	91.5 ± 9.6	102	40.5 ± 5.0	119	ND	132	91
Brn	0.589	260.4 ± 21.0	103	198.6 ± 10.0	86	76	128.5 ± 19.9	85	49.9 ± 8.4	108	ND	178.4	93
WMG	0.962	99.1 ± 4.4	108	81.3 ± 4.4	94	82	59.8 ± 2.3	83	19.8 ± 4.9	105	ND	79.6	97
WMG	3.518	118.8 ± 2.6	103	90.0 ± 1.3	85	76	72.3 ± 8.6	89	25.4 ± 1.4	108	ND	97.7	101
WMG	0.65	181.4 ± 3.2	101	134.0 ± 6.0	102	74	77.0 ± 5.5	93	56.4 ± 5.7	113	ND	133.4	98
WMG	3.706	166.4 ± 17.2	95	125.0 ± 18.5	98	75	59.5 ± 2.3	95	55.4 ± 5.3	97	ND	114.9	94
WMG	0.68	176.1 ± 16.0	103	97.1 ± 4.9	87	55	58.9 ± 0.5	92	40.4 ± 1.4	88	ND	99.3	92
WMG	6.384	73.8 ± 10.0	99	59.6 ± 1.2	91	81	48.5 ± 3.2	96	11.5 ± 2.0	91	ND	60	101
WMG	6.384	48.0 ± 2.8	96	38.9 ± 3.0	91	81	34.3 ± 1.6	88	8.4 ± 0.5	93	ND	42.7	102
WMG	1.186	195.9 ± 16.4	99	131.8 ± 7.9	88	67	71.1 ± 5.6	87	51.9 ± 5.9	86	ND	123	93
WLG	3.212	183.1 ± 3.4	93	120.3 ± 3.0	81	66	70.7 ± 5.8	97	43.9 ± 3.1	93	ND	114.6	95
WLG	3.212	210.9 ± 32.4	113	115.4 ± 15.2	84	55	62.0 ± 10.0	95	34.8 ± 1.7	85	ND	96.8	88
WLG	3.212	178.8 ± 14.2	109	117.4 ± 4.0	92	66	59.5 ± 4.3	90	46.9 ± 2.9	105	ND	106.4	89
WLG	3.212	181.9 ± 26.4	115	119.7 ± 23.5	95	66	64.0 ± 2.8	87	48.7 ± 0.7	102	ND	112.7	94
WLG	0.939	228.7 ± 10.4	96	160.1 ± 10.6	86	70	89.8 ± 2.9	87	63.8 ± 5.3	91	ND	153.6	96
WLG	8.042	186.2 ± 11.0	100	139.0 ± 5.4	96	75	56.2 ± 4.0	88	61.1 ± 8.5	113	ND	153.6	88
WLG	4.941	185.9 ± 10.2	97	142.4 ± 4.2	92	77	57.8 ± 1.7	79	66.2 ± 6.0	94	ND	124	89
WLG	5.683	166.7 ± 2.6	93	113.2 ± 7.3	92	68	45.2 ± 5.3	88	51.0 ± 2.0	89	ND	96.2	85
WLG	1.977	202.4 ± 25.4	88	144.4 ± 10.8[j]	82[j]	71[j]	64.1 ± 6.5	82	70.5 ± 6.2	96	ND	134.6	95
WLG	5.176	151.6 ± 16.0	101	100.7 ± 1.8	90	66	55.8 ± 3.7	87	35.8 ± 5.4	92	ND	91.6	92
WLG	7.144	151.1 ± 10.6	100	98.7 ± 6.8	82	65	52.9 ± 4.7	83	39.4 ± 2.6	97	ND	92.3	92
WLG	4.917	276.6 ± 22.2	82	197.1 ± 30.2[j]	89[j]	66[j]	71.3 ± 5.3	95	74.8 ± 11.7	120	9.7 ± 3.3	155.8	98
WLG	0.971	284.7 ± 20.6	99	166.2 ± 8.4	94	58	66.7 ± 1.3	95	81.8 ± 8.9	103	8.7 ± 1.2	163.2	88
Across Matrix Avg[k] ± 2σ		176.4 ± 116.5	100 ± 15	124.8 ± 76.4	90 ± 11	72 ± 16	71.1 ± 52.2	90 ± 10	39.6 ± 37.7	94 ± 21		115.5 ± 70.4	91 ± 12
SRM[l] 1568a ($\bar{X} \pm 2\sigma$)		299.1 ± 31.2		266.9 ± 41.2		90 ± 18	76.9 ± 16.1		162.3 ± 22.5			250.7 ± 39.3	95 ± 13

Abbreviations: As_{TD}, the total digested arsenic; As_{TE}, total extracted arsenic; $As_{\Sigma Chrom}$, the sum of the individual arsenic species in a rice composite; Brn, brown rice; DMA, dimethylarsinic acid; DMMTA, dimethylmonothioarsinic acid; iAs, inorganic arsenic; Ins, instant rice; LFB, laboratory-fortified blank; LFM, laboratory-fortified matrix; MMA, monomethylarsonic acid; ND, no detect; Par, parboiled rice; RWF, relative weighting factor; SRM, standard reference material; WLG, white long grain rice; WMG, white medium grain rice; WSG, white short grain rice.

[a]All analyses are reported based on oven-dried sample weights. All concentrations (ng/g) are reported as averages ($\bar{x} \pm 2\sigma$, $n = 3$ replicates from a composited cooked rice sample) except where noted. [b]Two types of total arsenic LFBs were analyzed. The first was a LFB As_{TD} and the second was a LFB As_{TE}. The LFB As_{TD} was 96 ± 14% and the LFB As_{TE} was 99 ± 11%. The samples were fortified prior to digestion with 4.8 to 8 ppb As for the As_{TD} LFB and LFM. Two types of species-specific arsenic LFBs were analyzed. The first was a LFB for the iAs (LFB_{iAs}) and the second was a LFB for DMA (LFB_{DMA}). The LFB_{iAs} was 97 ± 27% and the LFB_{DMA} was 99 ± 30%. The LFB extracts were fortified with 4 ppb iAs, 3 ppb DMA, and 1.5 ppb MMA. [c]Samples were categorized according to U.S. Rice Federation Production classifications. [d]RWF is calculated based on mill- and grain-specific production data relative to all the domestic rice intended for direct human consumption [(mill- and grain-specific production/all production intended for human consumption) × 100]. [e]As_{TD} is a total arsenic determination after a hot mineral acid dissolution. LFM As_{TD} is a LFM on a sample that will undergo a hot mineral acid dissolution. [f]As_{TE} is a flow injection total arsenic determination in the extraction fluid. As_{TE} is determined using a single-point method of standard addition. LFM As_{TE} is a LFM on the total arsenic in the extraction fluid. [g]Extraction efficiency equals the As_{TE} in a rice composite, divided by the total arsenic after a hot mineral acid dissolution (As_{TD}) times 100. [h]The LFM extract was fortified with 4 ppb iAs, 3 ppb DMA, and 1.5 ppb MMA. Inorganic arsenic (iAs = As^{III} + As^{V}). LFM_{iAs} is a species-specific LFM for iAs in the extraction fluid. LFM_{DMA} is a species-specific LFM for DMA in the extraction fluid. LFM_{MMA} is a species-specific LFM for MMA in the extraction fluid. [i]Average chromatographic recovery equals $As_{\sum Chrom}$ divided by As_{TE} for that rice composite times 100. [j]These concentrations and percentages were determined using $n = 2$. [k]Across-matrix average and sigma are calculated using the mean for each rice composite and do not include data from the SRM. The across-matrix average and sigma are arithmetic not production weighted. [l]The mass balance terms have been calculated for the SRM by pooling the data from individual batches ($n = 40$).

Table S6. Summary statistics of one-day rice consumption rates for the U.S. population and both ethnic- and age-based subpopulations.[a]

Consumption of Rice for U.S. Populations	n	Mean		50th Percentile		75th Percentile		90th Percentile		95th Percentile	
		g/day	$\frac{g}{kg\ BW\text{-}day}$	$\frac{\times 10^{-2}\ g}{day}$	$\frac{\times 10^{-3}\ g}{kg\ BW\text{-}day}$	g/day	$\frac{g}{kg\ BW\text{-}day}$	g/day	$\frac{g}{kg\ BW\text{-}day}$	g/day	$\frac{g}{kg\ BW\text{-}day}$
General Population											
Mexican American	14,009	16.74	0.320	0.55	0.108	21.16	0.386	56.42	1.030	83.42	1.529
Other Hispanic	2,009	31.91	0.565	707.17	149.391	49.37	0.831	90.68	1.619	119.38	2.286
Non-Hispanic White	19,239	12.74	0.197	0.55	0.084	9.95	0.158	44.77	0.667	71.78	1.072
Non-Hispanic Black	12,389	16.94	0.291	0.31	0.049	20.10	0.306	58.49	0.965	86.51	1.517
Tribal, Asian, and Pacific	2,136	41.63	0.790	234.10	36.874	59.95	1.112	123.44	2.419	169.26	3.461
All U.S.	49,782	15.72	0.269	0.57	0.101	14.92	0.252	55.97	0.872	84.63	1.414
1−2-year-olds											
Mexican American	1,062	8.15	0.652	1.26	0.930	11.98	0.982	28.26	2.282	41.20	3.281
Other Hispanic	154	15.39	1.311	686.89	551.823	20.25	2.045	44.18	4.186	61.82	5.013
Non-Hispanic White	925	4.82	0.386	0.36	0.288	1.44	0.107	14.71	1.272	26.25	2.248
Non-Hispanic Black	759	8.14	0.651	0.35	0.259	9.47	0.825	28.21	2.304	41.65	3.195
Tribal, Asian, and Pacific	155	19.74	1.545	1.61	1.260	27.14	2.281	62.51	5.324	90.59	7.515
All U.S.	3,055	7.03	0.581	0.56	0.474	5.40	0.476	22.41	1.990	41.20	3.288

Abbreviations: BW, body weight; iAs, inorganic arsenic.
[a]The exposure estimates for 25th percentile and below were all zero. Cooked rice assumes a rice-to-water ratio (v/v) of approximately 1:2. The *n* reported is associated with the individuals who reported their body weights. There is uncertainty in estimating the consumption rates associated with the upper and lower population percentiles because of the small sample size in the tails of the distribution and the use of 24-hour dietary recall questionnaire.

Table S7. Comparison of one-day and long-term rice consumption rate estimates in the U.S. population based on WWEIA short-term survey data and NHS, NHS II, and HPFS long-term survey data[a]

Survey[b]	Age of Subpopulation	Gender of Subpopulation	Ethnicity of Surveyed Population	n	Estimates are in Cooked Cups of Rice/day		
					Mean	90th Percentile	95th Percentile
WWEIA[c]	26-45	Female	Non-Hispanic White Only	2,240	0.19	0.74	1.12
NHS II[d]	26-45	Female	92% EA, 1.7% A, 1.5% AA & 4.3% O	96,734	0.17	0.43	0.50
WWEIA[c]	37-65	Female	Non-Hispanic White Only	2,763	0.16	0.56	1.03
NHS[d]	37-65	Female	97.7% EA, 0.7% A, 1.1% AA & 0.5% O	80,619	0.12	0.25	0.43
WWEIA[c]	26-84	Male	Non-Hispanic White Only	5,129	0.30	1.07	1.59
HPFS[d]	32-87	Male	94.5% EA, 1.7% A, 1.0% AA & 2.4% O	47,697	0.21	0.43	0.56

Abbreviations: WWEIA, What We Eat in America; NHS, Nurses' Health Study; HPFS, Health Professional Follow-Up Study; EA European Americans; A, Asian; AA, African-American; O, Other

a. Analyses of all four surveys utilized all participants to estimate the consumption rates (rather than limiting the analysis to the participants who reported rice consumption). These comparisons across studies are limited by differences between the survey questionnaires (i.e., long-term vs. short-term recall) and the positive bias produced by the inclusion of indirect (rice as an ingredient in processed foods) rice consumption within the WWEIA survey, which is not captured by the NHS, NHS II and HPFS surveys.
b. Rice consumption rates estimated from: WWEIA from 2001 - 2006; NHS II years 2003 & 2007; NHS years 2002 & 2006; and HPFS years 2002 & 2006.
c. WWEIA store bought rice consumption rate were converted to cups of cooked rice by converting grams of store bought to grams of oven dried rice (store bought rice is assumed to be 11% water by weight); converting grams of oven dried rice to grams of cooked rice (cooked rice is assumed to be 70% water by weight) and finally converting grams of cooked rice to cups of cooked rice (1 cup cooked rice is assumed to correspond to 158 grams of cooked rice)(USDA 2015a)
d. These data were graciously provided by Dr. Qi Sun and Dr Gang Liu from the Department of Nutrition, Harvard T.H. Chan School of Public Health, Boston, MA and Dr. Xuehong Zhang from the Department of Medicine, Channing Division of Network Medicine, Brigham and Women's Hospital and Harvard Medical School, Boston, Ma.

Short-term rice consumption surveys cannot be used reliably to predict long-term rice consumption rates. To compare long-term rice consumption rate estimates relative to the short-term survey data, Table S7 compares single-day (direct + indirect) rice consumption estimates for specific age, gender and ethnically mixed WWEIA subpopulations to longer term daily (direct) rice consumption rates estimated using the longer term questionnaires associated with the Nurses' Health Study, the Nurses' Health Study II and the Health Professional Follow-Up Study (Sun, Q, Liu, G, Zhang, X, unpublished data). Specifically, the three studies asked the following questions regarding direct white (1 cup) and brown (1 cup) rice consumption: "For each food listed, fill in the circle indicating how often on average you have used the amount specified during the past year." The answer can be given in 9 incremental choices from "Never, or less than once a month" to "6+ per day". By comparison, WWEIA estimates direct and indirect rice consumption via a two day 24-hour dietary recall. The first day involves an interview conducted in person in the Mobile Examination

Center and the second is conducted by telephone using the Automated Multiple-Pass Method. Although this manuscript does not develop long-term rice consumption rates or long-term iAs intake rate estimates based on rice consumption rates, there is public health interest in developing such estimates (Zhang et al. 2016).

Table S8. Summary statistics of iAs exposure from cooked rice using a gastrointestinal extraction procedure for the U.S. population and both ethnic- and age-based subpopulations.[a]

Exposure to iAs of Rice for Various Populations	*n*	Mean		50th Percentile		75th Percentile		90th Percentile		95th Percentile	
		µg/day	µg/kg BW-day	× 10⁻² µg/day	× 10⁻³ µg/kg BW-day	µg/day	µg/kg BW-day	µg/day	µg/kg BW-day	µg/day	µg/kg BW-day
General Population											
Mexican American	14,009	1.13	0.020	0.05	0.009	1.41	0.023	3.74	0.063	5.61	0.097
Other Hispanic	2,009	2.11	0.034	52.31	7.926	3.16	0.050	5.96	0.097	8.20	0.142
Non-Hispanic White	19,239	0.87	0.013	0.05	0.007	0.71	0.010	3.01	0.042	4.87	0.067
Non-Hispanic Black	12,389	1.15	0.019	0.03	0.004	1.33	0.018	3.89	0.059	5.86	0.094
Tribal, Asian, and Pacific	2,136	2.77	0.049	16.84	2.175	3.83	0.067	8.13	0.147	11.73	0.215
All U.S.	49,782	1.09	0.017	0.05	0.008	1.07	0.015	3.65	0.054	5.79	0.087
1−2-year-olds											
Mexican American	1,062	0.57	0.042	0.10	0.070	0.74	0.057	2.02	0.142	2.77	0.218
Other Hispanic	154	1.09	0.088	49.93	44.553	1.49	0.133	3.08	0.246	3.86	0.331
Non-Hispanic White	925	0.40	0.031	0.03	0.023	0.09	0.007	1.10	0.083	2.10	0.157
Non-Hispanic Black	759	0.58	0.044	0.03	0.023	0.66	0.051	2.01	0.149	2.97	0.219
Tribal, Asian, and Pacific	155	1.31	0.099	0.15	0.112	1.43	0.122	3.76	0.334	6.37	0.502
All U.S.	3,055	0.55	0.042	0.05	0.035	0.47	0.032	1.71	0.133	2.97	0.217

Abbreviations: BW, body weight; iAs, inorganic arsenic.
[a]The exposure estimates for 25th percentile and below were all zero. Cooked rice assumes a rice-to-water ratio (v/v) of approximately 1:2. The *n* reported is associated with the individuals who reported their body weights. There is uncertainty in estimating the consumption and exposure rates associated with the upper and lower population percentiles because of the use of the single 24-hour dietary recall and added uncertainty associated with small sample size.

Table S9. Summary statistics of iAs exposure from cooked rice using the DNAS procedure for the U.S. population and both ethnic- and age-based subpopulations.[a]

Exposure to iAs of Rice for Various Populations	n	Mean		50th Percentile		75th Percentile		90th Percentile		95th Percentile	
		μg/day	μg / kg BW-day	× 10^{-2} μg / day	× 10^{-3} μg / kg BW-day	μg/day	μg / kg BW-day	μg/day	μg / kg BW-day	μg/day	μg / kg BW-day
General Population											
Mexican American	14,009	1.59	0.031	0.06	0.012	1.97	0.035	5.33	0.097	8.01	0.148
Other Hispanic	2,009	2.98	0.053	72.26	12.942	4.57	0.075	8.80	0.152	11.68	0.217
Non-Hispanic White	19,239	1.24	0.019	0.06	0.009	0.96	0.014	4.31	0.064	6.80	0.104
Non-Hispanic Black	12,389	1.62	0.028	0.04	0.006	1.84	0.027	5.61	0.090	8.56	0.148
Tribal, Asian, and Pacific	2,136	3.92	0.075	24.30	3.735	5.51	0.102	12.02	0.228	16.82	0.321
All U.S.	49,782	1.55	0.026	0.07	0.011	1.49	0.023	5.23	0.082	8.20	0.136
1−2-year-olds											
Mexican American	1,062	0.77	0.061	0.15	0.104	1.11	0.083	2.65	0.209	3.75	0.305
Other Hispanic	154	1.48	0.126	60.36	57.337	2.21	0.198	4.48	0.360	5.16	0.443
Non-Hispanic White	925	0.50	0.040	0.04	0.032	0.12	0.009	1.47	0.122	2.66	0.215
Non-Hispanic Black	759	0.79	0.064	0.04	0.029	0.96	0.076	2.86	0.228	4.15	0.324
Tribal, Asian, and Pacific	155	1.89	0.148	0.19	0.163	2.70	0.243	5.82	0.410	8.14	0.654
All U.S.	3,055	0.72	0.057	0.07	0.055	0.58	0.045	2.32	0.193	3.80	0.296

Abbreviations: BW, body weight; DNAS, dilute nitric acid extraction; iAs, inorganic arsenic.
[a]The exposure estimates for 25th percentile and below were all zero except for 1−2-year-old Other Hispanic which was 0.04 × 10^{-2} μg/day (0.04 × 10^{-3} μg/kg BW-day) and for 1−2-year-old Tribal, Asian, and Pacific which was 0.01 × 10^{-2} μg/day (0.004 × 10^{-3} μg/kg BW-day). Cooked rice assumes a rice-to-water ratio (v/v) of approximately 1:2. The n reported is associated with the individuals who reported their body weights. There is uncertainty in estimating the exposure rates associated with the upper and lower population percentiles because of the use of the single 24-hour dietary recall and added uncertainty associated with small sample size.

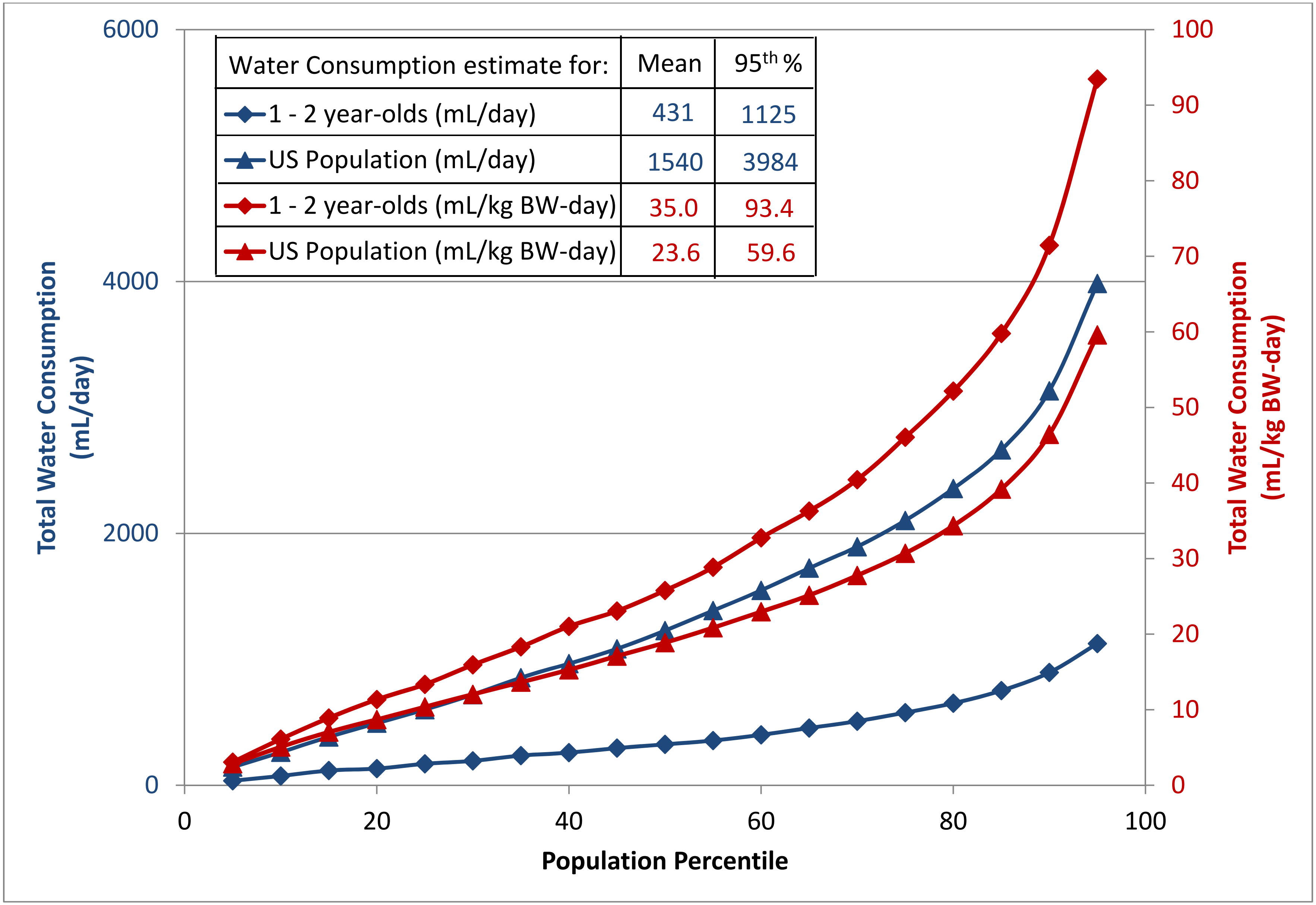
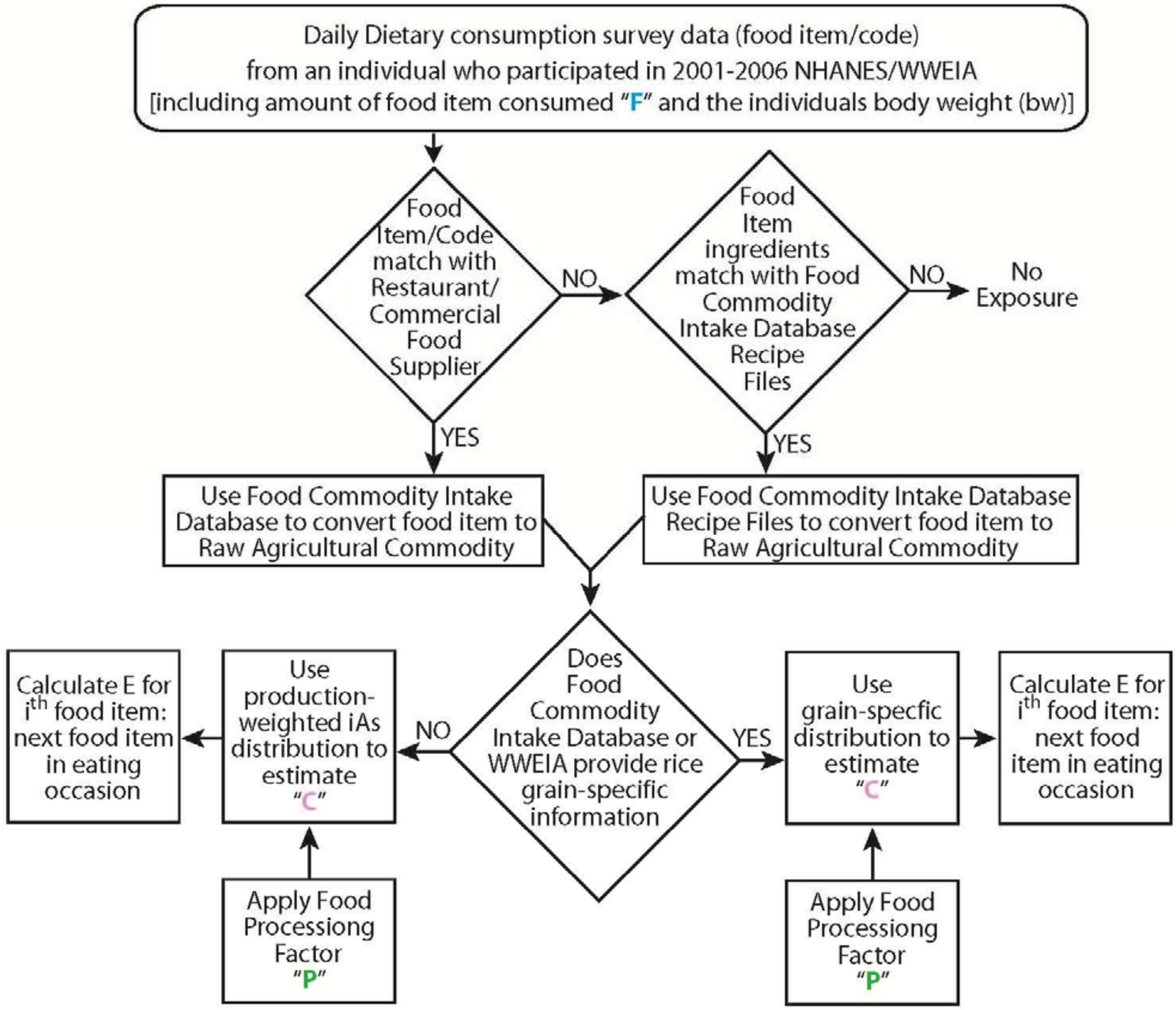


Figure S1. Cumulative Density Function Plots for total water consumption (mL/day and mL/kg BW-day) for U.S. population and children 1−2 years old.[a]

Abbreviations: BW, body weight.

[a]Total water comes from three sources, which include direct + indirect (food prep.) + other fluids (bottled water, etc.). There is uncertainty in estimating the daily water consumption rates associated with the lower and upper population percentiles because of the use of the single 24-hour dietary recall.



$$E_{EO} = \sum_{i=1}^{n} F \times C \times P$$

where:

- E_{EO} = Individual's Dietary Exposure for a Single Eating Occasion (µg chemical/ kg bw)
- F = Amount of Food item consumed (g food/kg bw)
- C = Concentraion in the food item (µg chemical/g food)
- P = procession factor (unitless)
- n = number of unique foods (or food commodities) consumed per eating occasion

$$TE = \sum_{j=1}^{n} E_{EO}$$

where:

- TE = Individual's Total Daily Exposure (µg chemical/ kg bw)
- E_{EO} = Individual's Dietary Exposure for a Single Eating Occasion (µg chemical/ kg bw)
- n = Number of eating occasions in a given day.

Figure S2. Flowchart and equations used by the SHEDS model to convert WWEIA food items into Food Commodity Intake Database Raw Agriculture Commodities used to estimate iAs exposures.

Abbreviations: iAs, inorganic arsenic; SHEDS, Stochastic Human Exposure and Dose Simulation; WWEIA, What We Eat in America.

Supplemental Material, Figure S2 is a flowchart outline of the process used by the Stochastic Human Exposure and Dose Simulation (SHEDS) model to produce an individual's iAs exposure from rice associated with a single food item/code within an eating occasion. The model takes the individual food codes and dietary consumption data from an individual's single eat occasion within the National Health and Nutrition Examination Survey /What We Eat in America (WWEIA) 2001−2006 and attempts to match the item with a restaurant/commercial food supplier's product within the U.S. EPA/USDA Food Commodity Intake Database recipe files. If an exact match is found, the database is used to directly convert the food item into a raw agricultural commodity. For example, food code 57308400 is a multigrain cereal, which has been translated by Food Commodity Intake Database into the following list of Raw Agriculture Commodities.

Recipe for multigrain cereal:

Raw Agriculture Commodity	Percent	Raw Agriculture Commodity	Percent
Barley, pearled barley	9.66	**Rice, brown**	**9.66**
Beet, sugar	12.75	Sugarcane, sugar	16.23
Corn, field, meal	14.21	Wheat, bran	9.66
Cottonseed, oil	1.30	Wheat, flour	9.66
Oat, groats/rolled oats	9.66	Wheat, grain	9.66

In this example, 9.66% of the consumption is from brown rice. In this case, the Food Commodity Intake Database database provides the grain-specific information, and SHEDS uses a brown rice-only distribution to estimate the iAs concentration (*C*) in this commodity for this food item/code associated with this eating occasion. This information is combined with a food-processing factor (*P*, concentration or dilution factors due to processes of food from the Raw Agriculture Commodity into food products) and the amount consumed (*F*) to estimate the iAs exposure from the rice associated with that food item/code for that eating occasion. However, if an exact food item/code is not found, then the recipe files within Food Commodity Intake Database attempt to convert it into its corresponding Raw Agriculture Commodity. For example, a rice, fried with poultry meat is translated into the following list of Raw Agriculture Commodities.

Recipe for Rice, Fried with Meat, Poultry:

Raw Agriculture Commodity	Percent	Raw Agriculture Commodity	Percent
Bean, mung, seed	0.49	Rapeseed, oil	0.17
Chicken, fat	0.50	**Rice, white**	**24.24**
Chicken, meat	6.29	Safflower, oil	<0.01
Corn, field, oil	0.20	Sesame, oil	<0.01
Cottonseed,oil	0.32	Soybean, oil	3.56
Egg, whole	10.74	Soybean, seed	0.96
Olive, oil	0.08	Sunflower, oil	0.01
Onion, green	2.80	Water, indirect, all sources	39.31
Pea, succulent	3.42	Wheat, flour	0.38
Peanut, oil	0.06		

In this case, 24.24% of the consumption is from white rice. Again, the grain-specific information is provided, and SHEDS combines this with the *P,* and *F* factors to estimate the iAs exposure from the rice associated with that food code and eating occasion.

In either of the above examples, if the grain-specific information was not provided, an iAs concentration for the rice component of the Raw Agriculture Commodity would be estimated by using a production-weighted distribution. The exposure for a particular eating occasion is calculated by summing all the iAs exposure from each food item/code within that eating occasion. The summation of exposures from every eating occasion within one day yields the individual's total daily exposure. Finally, the equations and a definition of associated terms is provided at the bottom of Supplemental Material, Figure S2.

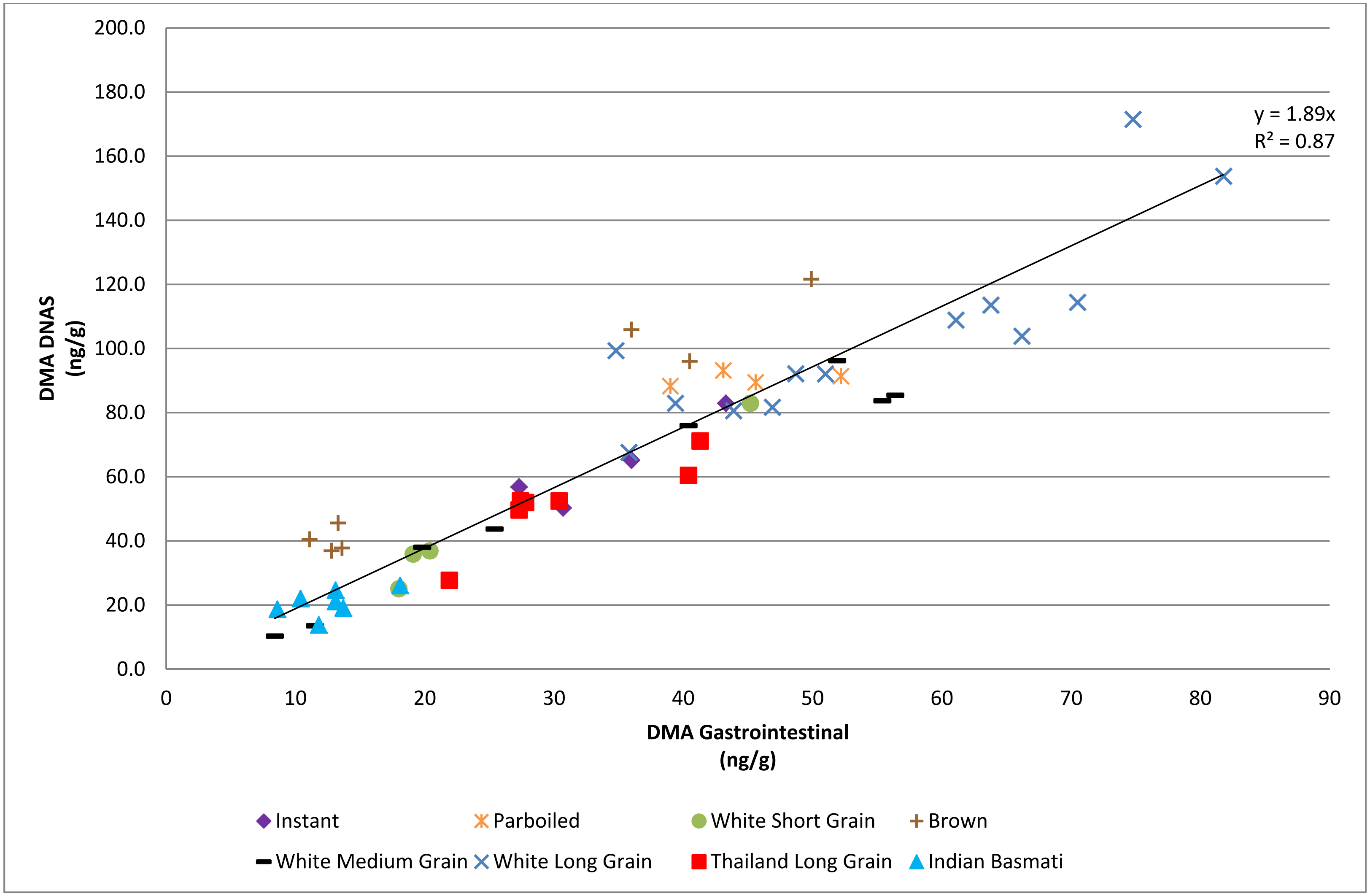


Figure S3. Correlation of grain-specific DMA concentrations in U.S.-consumed rice determined after DNAS and gastrointestinal-based extractions.

Abbreviations: DMA, dimethylarsinic acid; DNAS, dilute nitric acid extraction.

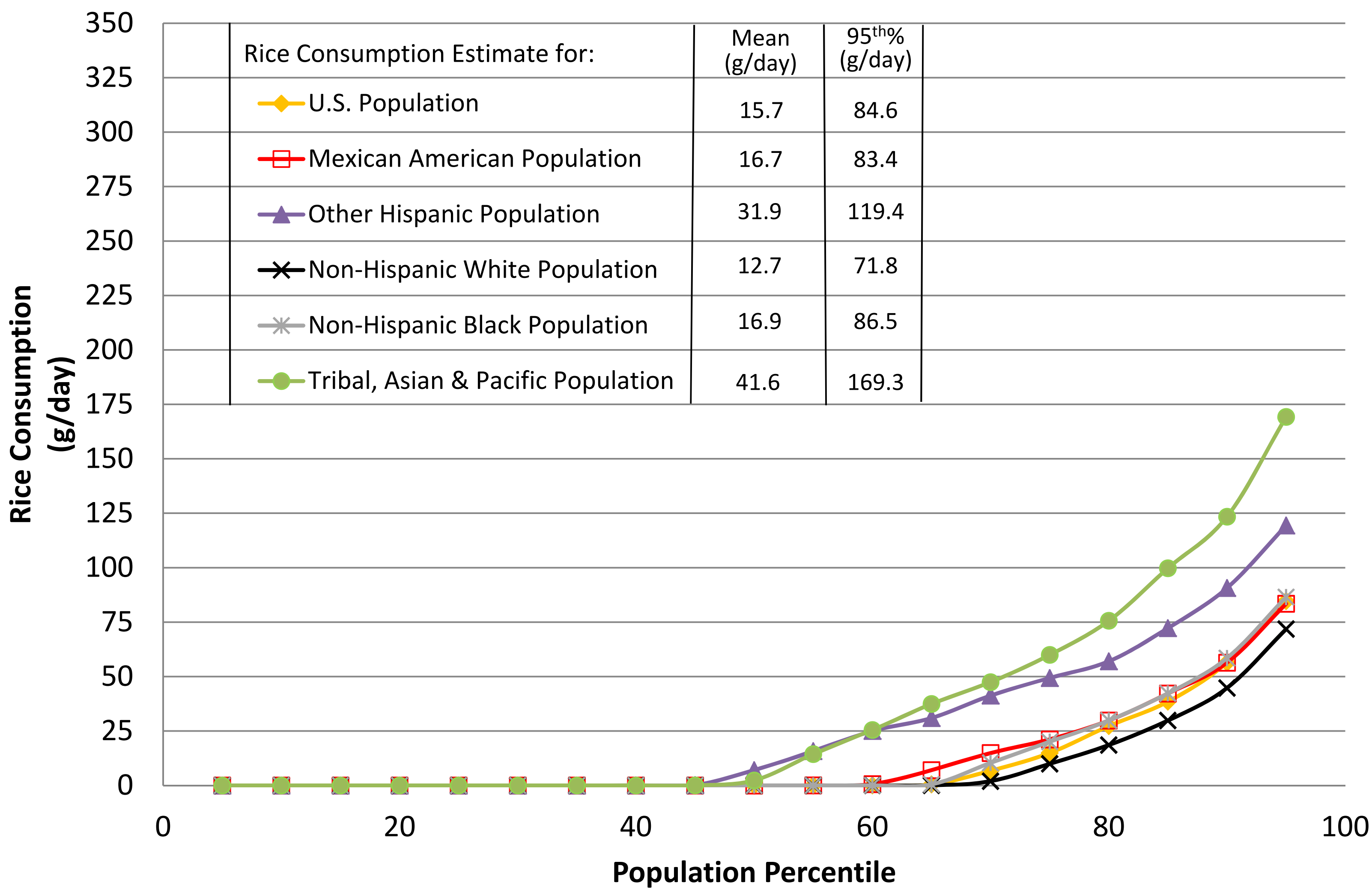


Figure S4. Cumulative Density Function Plots of rice consumption (g/day) for U.S. population and ethnic subpopulations.[a]

[a]There is uncertainty in estimating the rice consumption rates associated with the upper and lower population percentiles because of the use of the single 24-hour dietary recall and added uncertainty associated with small sample sizes.

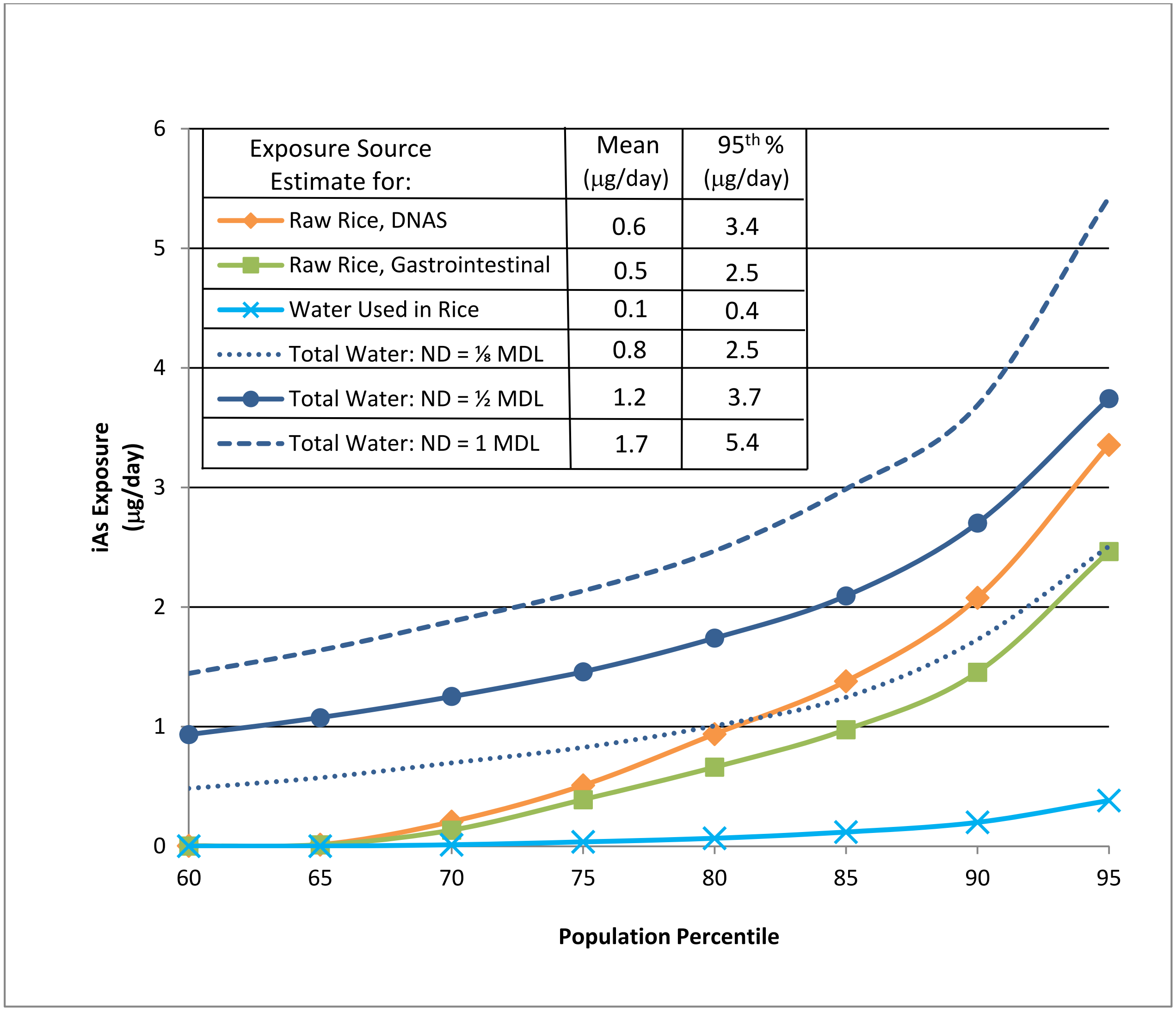


Figure S5. Cumulative Density Function Plots of iAs exposure (µg/day) from rice, water used to cook rice, and total water consumption for the 1−2-year-old U.S. subpopulation.[a]

Abbreviations: iAs, inorganic arsenic; MDL, method detection limit; ND, no detect.

[a]Total water comes from three sources, which include direct + indirect (food prep) + other fluids (bottle water, etc). There is uncertainty in estimating the iAs exposure rates associated with the upper and lower population percentiles because of the use of the single 24-hour dietary recall.

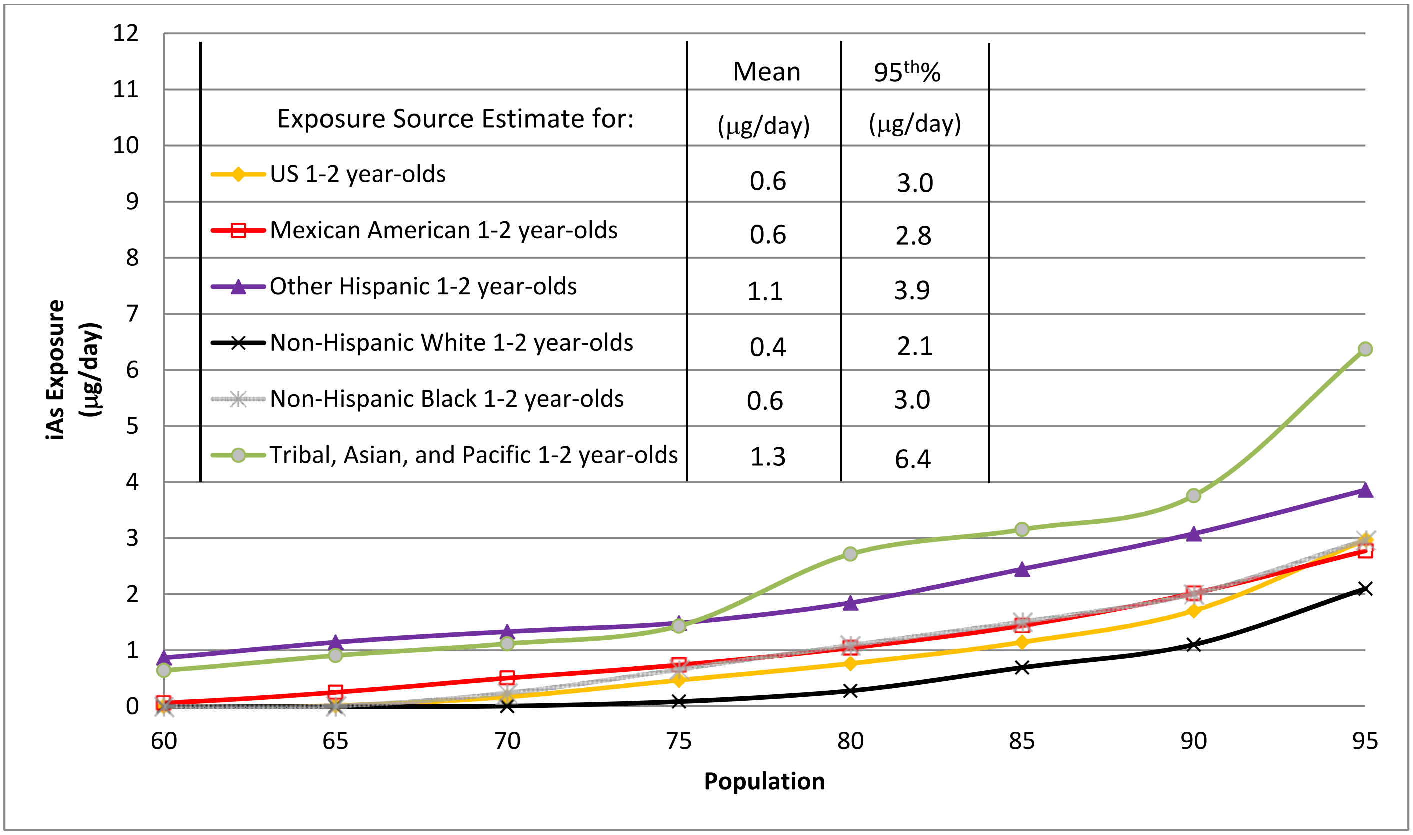


Figure S6. Cumulative Density Function Plots of iAs exposures (µg/day) from cooked rice for 1−2-year-old children from different ethnicities using a gastrointestinal extraction procedure.[a]

Abbreviations: iAs, inorganic arsenic.
[a]There is uncertainty in estimating the iAs exposure rates associated with the upper population percentiles because of the use of the single 24-hour dietary recall and added uncertainty associated with small sample size.

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